

10/11/00


jc803 U.S. PTO


jc921 U.S. PTO

09/85341

10/11/00

UTILITY PATENT APPLICATION TRANSMITTAL <small>(Only for new nonprovisional applications under 37 CFR 1.53(b))</small>	Attorney Docket No.	198277US0 DIV
	First Inventor or Application Identifier	Peter JENKNER
	Title	FLUOROALKYL-FUNCTIONAL ORGANOSILOXANE-CONTAINING COMPOSITIONS BASED ON ALCOHOL, A PROCESS FOR THEIR PREPARATION AND THEIR USE

APPLICATION ELEMENTS <small>See MPEP chapter 600 concerning utility patent application contents</small>	ADDRESS TO: Assistant Commissioner for Patents Box Patent Application Washington, DC 20231
<p>1. <input checked="" type="checkbox"/> Fee Transmittal Form (e.g. PTO/SB/17) (Submit an original and a duplicate for fee processing)</p> <p>2. <input checked="" type="checkbox"/> Specification Total Pages 19</p> <p>3. <input type="checkbox"/> Drawing(s) (35 U.S.C. 113) Total Sheets <input type="text"/></p> <p>4. <input checked="" type="checkbox"/> Oath or Declaration Total Pages 4</p> <p>a. <input type="checkbox"/> Newly executed (original or copy)</p> <p>b. <input checked="" type="checkbox"/> Copy from a prior application (37 C.F.R. §1.63(d)) (for continuation/divisional with box 15 completed)</p> <p>i. <input type="checkbox"/> DELETION OF INVENTOR(S) Signed statement attached deleting inventor(s) named in the prior application, see 37 C.F.R. §1.63(d)(2) and 1.33(b).</p> <p>5. <input checked="" type="checkbox"/> Incorporation By Reference (usable if box 4B is checked) The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4B, is considered to be part of the disclosure of the accompanying application and is hereby incorporated by reference therein.</p>	ACCOMPANYING APPLICATION PARTS <p>6. <input checked="" type="checkbox"/> The prior application is assigned of record to: <u>Huels Aktiengesellschaft, Recordation Date: 08/21/1998, Reel/Frame: 9406/0755-0757.</u></p> <p>7. <input type="checkbox"/> 37 C.F.R. §3.73(b) Statement (when there is an assignee) <input type="checkbox"/> Power of Attorney</p> <p>8. <input type="checkbox"/> English Translation Document (if applicable)</p> <p>9. <input checked="" type="checkbox"/> Information Disclosure Statement (IDS) <input type="checkbox"/> Copies of IDS Citations</p> <p>10. <input checked="" type="checkbox"/> Preliminary Amendment</p> <p>11. <input checked="" type="checkbox"/> White Advance Serial Number Postcard</p> <p>12. <input type="checkbox"/> Small Entity Statement(s) <input type="checkbox"/> Statement filed in prior application. Status still proper and desired.</p> <p>13. <input type="checkbox"/> Certified Copy of Priority Document(s) (if foreign priority is claimed)</p> <p>14. <input checked="" type="checkbox"/> Other: Request for Priority; Form PTO-1449</p>
<p>15. If a CONTINUING APPLICATION, check appropriate box, and supply the requisite information below:</p> <p><input type="checkbox"/> Continuation <input checked="" type="checkbox"/> Divisional <input type="checkbox"/> Continuation-in-part (CIP) of prior application U.S. Serial Number <u>08/984,162</u></p> <p>Prior application information: Examiner: P. Shaver Group Art Unit: 1621</p>	
<p>16. Amend the specification by inserting before the first line the sentence:</p> <p><input checked="" type="checkbox"/> This application is a <input type="checkbox"/> Continuation <input checked="" type="checkbox"/> Divisional <input type="checkbox"/> Continuation-in-part (CIP) of prior application U.S. Serial Number <u>08/984,162</u>, filed on <u>December 3, 1997</u>.</p> <p><input type="checkbox"/> This application claims priority of provisional application Serial Number _____, filed on _____.</p>	
<p>17. CORRESPONDENCE ADDRESS</p> <p></p> <p>22850 (703) 413-3000 FACSIMILE: (703) 413-2220</p>	

Name:	Norman F. Oblon	Registration No.:	24,618
Signature:		Date:	10/11/00
Name:	Frederick D. Vastine, Ph.D.	Registration No.:	27,013

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INVENTORS: Peter JENKNER et al

SERIAL NO.: NEW APPLICATION

FILING DATE: HEREWITH

FOR: FLUOROALKYL-FUNCTIONAL ORGANOSILOXANE-CONTAINING COMPOSITIONS
BASED ON ALCOHOL, A PROCESS FOR THEIR PREPARATION AND THEIR USEjc921 U.S. PTO
09/685341
10/11/00**FEE TRANSMITTAL**ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

FOR	NUMBER FILED	NUMBER EXTRA	RATE	CALCULATIONS
TOTAL CLAIMS	16 - 20 =	0	× \$18 =	\$0.00
INDEPENDENT CLAIMS	8 - 3 =	5	× \$80 =	\$400.00
<input type="checkbox"/> MULTIPLE DEPENDENT CLAIMS (If applicable)			+ \$270 =	\$0.00
<input type="checkbox"/> LATE FILING OF DECLARATION			+ \$130 =	\$0.00
BASIC FEE				\$710.00
TOTAL OF ABOVE CALCULATIONS				\$1,110.00
<input type="checkbox"/> REDUCTION BY 50% FOR FILING BY SMALL ENTITY				\$0.00
<input type="checkbox"/> FILING IN NON-ENGLISH LANGUAGE			+ \$130 =	\$0.00
<input type="checkbox"/> RECORDATION OF ASSIGNMENT			+ \$40 =	\$0.00
TOTAL				\$1,110.00

- ☐ Please charge Deposit Account No. 15-0030 in the amount of \$_____. A duplicate copy of this sheet is enclosed.
- ☒ A check in the amount of \$1,110.00 to cover the filing fee is enclosed.
- ☒ The Commissioner is hereby authorized to charge any additional fees which may be required for the papers being filed herewith and for which no check is enclosed herewith, or credit any overpayment to Deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.

Respectfully Submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.*FD Vastine*

Norman F. Oblon

Registration No. 24,618

Frederick D. Vastine, Ph.D.

Registration No. 27,013

Date: 10/11/00**22850**Tel. (703) 413-3000
Fax. (703) 413-2220
(OSMMN 11/98)

DOCKET NO.: 198277US0 DIV

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: :

Peter JENKNER et al :

SERIAL NO.: NEW APPLICATION :

FILED: HEREWITH :

FOR: FLUOROALKYL-FUNCTIONAL ORGANOSILOXANE-
CONTAINING COMPOSITIONS BASED ON ALCOHOL,
A PROCESS FOR THEIR PREPARATION AND THEIR
USE

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

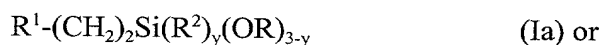
SIR:

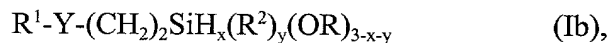
In advance of prosecution, please amend the above-identified application as follows.

IN THE CLAIMS

Please cancel Claims 1-22 in favor of the following new claims:

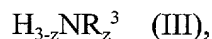
--23. A method of hydrophobizing and oleophobizing and for simultaneously providing a dirt- and color-repellent treatment of surfaces, of plastics, of metals, of textiles, leather, cellulose and starch products, and of mineral building materials, by applying to such an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:





in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ C$ over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1.

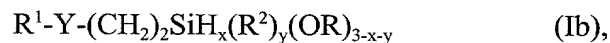
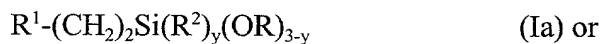
24. The method of Claim 23, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

25. A method of protecting buildings and facades, comprising:

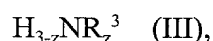
applying an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each

independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^{\circ}\text{C}$ over a period of $0.5-24$ hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of $2-500:1$, to buildings and facades.

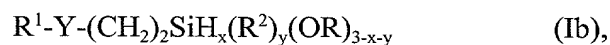
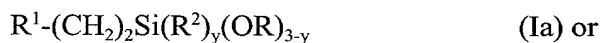
26. The method of Claim 25, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

27. A method for coating glass fibers, comprising:

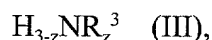
coating said glass fibers with an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^{\circ}\text{C}$ over a

period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1.

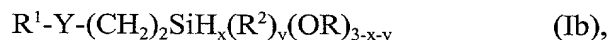
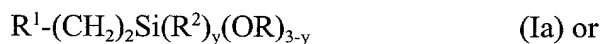
28. The method of Claim 27, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

29. A method of silanizing fillers and pigments, comprising:

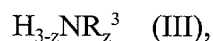
applying an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ\text{C}$ over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a

molar ratio of 2-500:1, to said fillers and pigments.

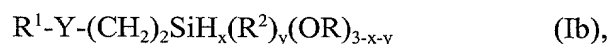
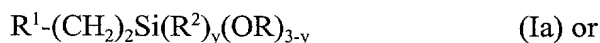
30. The method of Claim 29, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

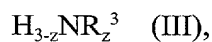
31. A method of improving the rheological properties of polymer dispersions and emulsions, comprising:

preparing said dispersions and emulsions with an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ C$ over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1.

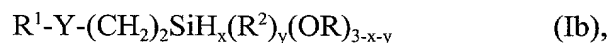
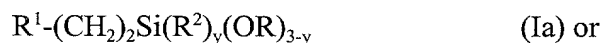
32. The method of Claim 31, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

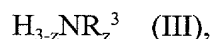
33. A method of providing a release layer with release properties, comprising:

incorporating an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ\text{C}$ over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1 in the release layer.

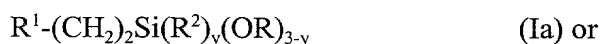
34. The method of Claim 33, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R³ is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, z = 1, 2 or 3 and groups R³ are identical or different.

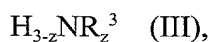
35. A method of formulating paints and coatings, comprising:

incorporating an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R¹ is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH₂, O or S group, R² and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and x = 0, 1 or 2 and y = 0, 1 or 2, where (x+y) ≤ 2, at a temperature in the range of 0-120°C over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1 in said paint or coating.

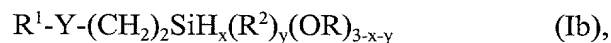
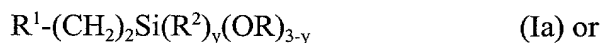
36. The method of Claim 35, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R³ is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, z = 1, 2 or 3 and groups R³ are identical or different.

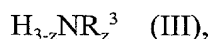
37. A method for promoting adhesion of a formulation, comprising:

incorporating an alcoholic fluoroalkyl-functional group containing organosiloxane based composition, which is essentially chlorine free, prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula Ia or Ib:



in which R¹ is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH₂, O or S group, R² and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and x = 0, 1 or 2 and y = 0, 1 or 2, where (x+y) ≤ 2, at a temperature in the range of 0-120°C over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and (1) a weak mono- or polybasic acid or (2) a weak base or (3) a weak mono- or polybasic acid and a weak base or (4) an acidic or basic salt, the water and alkoxysilane employed being in a molar ratio of 2-500:1 into said formulation.

38. The method of Claim 37, wherein said weak base of (2) and (3) is an alkyl amine of formula (III):



wherein R³ is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, z = 1, 2 or 3 and groups R³ are identical or different.--

REMARKS

Claims 1-22 have been canceled. New Claims 23-38 are active in the case.

Claims 15-22, restricted from the parent application U.S. Serial Number 08/984,162, are now pursued in this divisional application in the form of the new claims. Basis for the claims can be found in the written record of the claims in the parent application. No new matter has been introduced into the case.

It is believed that the claims are in proper condition for consideration.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.



Norman F. Oblon
Attorney of Record
Registration No.: 24,618

Frederick D. Vastine, Ph.D.
Registration No.: 27,013



22850

TEL: 703-413-3000
FAX: 703-413-2220

DOCKET NO.: 0689-0727-0

TITLE OF THE INVENTION

FLUOROALKYL-FUNCTIONAL ORGANOSILOXANE-CONTAINING
COMPOSITIONS BASED ON ALCOHOL, A PROCESS FOR
THEIR PREPARATION AND THEIR USE

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to fluoroalkyl-functional organopolysiloxane-containing compositions based on alcohol, to a process for their preparation and to their use.

Description of the Background

Organosilanes of the formula $R'-Si(R'')_3$, where R' is a fluorinated organic radical and R'' is a chlorine or methoxy or ethoxy group, have diverse uses, for example, in the application of hydrophobically and oleophobically acting layers on surfaces. Such coatings and impregnations can be produced at the worksite, for example, by dipping, impregnating, spraying or rolling operations.

It is also known that oil- and water-repellent coatings on surfaces, usually glass surfaces, can be produced with fluoroalkylalkoxysilanes. The coatings can be used, for example, as a dirt-repellent treatment on plate glass. The processes described are based on sol-gel processes, very fine inorganic particles being produced and employed together with the fluoroalkylalkoxysilane. Application of such systems is technically demanding, and usually uses complex organic solvent mixtures and additives. Moreover, such systems comprise chlorine and furthermore only remain fit to use for a

relatively short period of time (European Laid-Open Application No. 0 658 525, European Laid-Open Application No. 0 629 673 and U.S. 5 424 130).

Since the processes employed are sol-gel process
5 embodiments, which are demanding both in their use and
economically, their commercial utilization has to date been
limited to niche areas. Furthermore, there are narrow time
limits with respect to processability and, therefore, the
subsequent efficacy of such fluoroalkylsilane containing
10 formulations on a substrate surface. These factors are an
impediment to broader use of the formulation in practice. In
addition, a partial to complete loss of adhesive action can be
experienced, and a slow phase separation up to laminar
deposition of a fluoroalkylsilane-containing phase can occur.

15 The complicated application processes and, also the use
of solvents such as fluorinated hydrocarbons or
fluorohydrocarbons, therefore, are disadvantages of such
processes (European Laid-Open Application No. 0 491 251,
European Laid-Open Application No. 0 493 747).

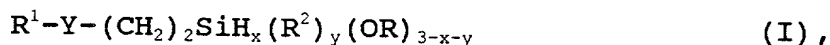
20 In the case of the known solvent-based systems, it must
be noted that although some systems are stable over the long
term in the non-hydrolyzed state, they are distinguished by an
efficacy limited to a maximum of a few days in the hydrolyzed
form because of the progressive polycondensation of the
25 silanol functions. When formulations are prepared in which
the content of silanol groups is continuously reduced in order
to reduce the undesired polycondensation, the extent of
chemical bonding by the silanol groups to a substrate surface

is furthermore reduced, this subsequently manifesting itself in unsatisfactory adhesion of the coating to the substrate surface. A need, therefore, continues to exist for an aqueous organosilane solution which is stable over prolonged periods of time and which provides coated substrates with both hydrophobic and oleophobic properties.

Summary of the Invention

Accordingly, one object of the present invention is to provide aqueous organosilane formulations which can be prepared in a simple and economical manner and which are essentially free of chlorine, and furthermore have adequate stability over relatively long periods of time, and which simultaneously impart hydrophobic and oleophobic properties on substrate surfaces by an impregnation process which is easy to carry out.

Briefly, this object and other objects of the present invention as hereinafter will become more readily apparent can be attained by an alcoholic fluoroalkyl-functional group containing organosiloxane based composition prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula I:

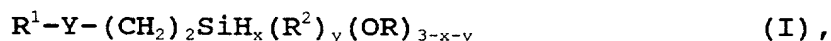


in which R^1 is a mono, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or

2, where $(x+y) \leq 2$, at a temperature in the range of 0-120°C over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which, contains water, and a weak mono- or polybasic acid or a weak base or a weak mono- or polybasic acid and a weak base or an acid or basic salt, the water employed and the alkoxysilane employed being in a molar ratio of 2-500:1.

Detailed Description of the Preferred Embodiments

It has been found, surprisingly, that aqueous organosiloxane-containing compositions which contain Si-bonded fluoroalkyl functional groups are accessible in a simple and economical manner as chlorine-free, homogeneous, clear solutions which are stable for several months if at least one fluoroalkyl-functional group containing organosilane of formula I:

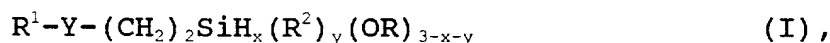


in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, is hydrolyzed in a controlled manner, with thorough mixing, in an alcoholic medium which, in addition to water, comprises a weak mono- or polybasic acid or a weak base or a weak mono or polybasic acid and a weak base or an acid or basic salt, at a temperature in the range of 0-120°C and over a period of 0.5-24 hours, the water and the alkoxysilane of

formula I being employed in a molar ratio of 2-500:1. The compositions of the invention furthermore can be employed for simultaneous hydrophobization and oleophobicization of substrate surfaces in a simple manner with an outstanding action.

5 Compositions of the invention preferably have a pH of 2-12, particularly preferably a pH of 3-10, and normally have a viscosity of less than 10,000 mPa·s. The content of alcohols in the compositions of the invention is preferably 40-99.999% by weight, based on the total composition, the composition
10 preferably comprising fluoroalkylfunctional organosiloxanes in amounts of 0.001-30% by weight, particularly preferably 0.01-5% by weight, and especially preferably 0.1-2% by weight, based on the composition.

Another aspect of the present invention relates to a
15 process for the preparation of a fluoroalkyl-functional organosiloxane containing composition, which comprises subjecting at least one fluoroalkyl-functional organosilane of formula I:



20 in which R^1 is a mono-, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or
25 2 , where $(x+y) \leq 2$, to controlled hydrolysis with thorough mixing, in an alcoholic medium which, in addition to water, comprises a weak mono- or polybasic acid or a weak base or a weak mono or polybasic acid and a weak base or an acid or

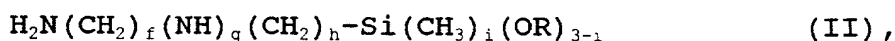
basic salt, at a temperature in the range of 0-120°C and over a period of 0.5-24 hours, preferably 1-12 hours, particularly preferably over a period of 2-6 hours, the water and the alkoxysilane being employed in a molar ratio of 2-500:1.

5 In the process of the invention, the fluoroalkyl-functional group containing organosilane of formula I is preferably metered into the reacting medium in portions with periodic interruptions. However, the metering operation can also be carried out continuously with periodic interruptions, 10 or the batchwise and continuous procedures for the metering can be combined with one another in a suitable manner.

The reaction is in general carried out at a temperature within the range of 0-120°C, preferably in a temperature range of 10-80°C, particularly preferably of 20-60°C. The reaction 15 medium is normally stirred.

In the process according to the invention, the pH of 2-12 in the reaction medium is suitably established by employing a weak mono- or polybasic acid or a weak base or a weak mono- or polybasic acid and a weak base or an acid or basic salt.

20 The weak base employed in the process of the invention is an organosilane of formula II:



in which $0 \leq f \leq 6$, $g=0$ if $f=0$ and $g=1$ if $f>1$, $0 \leq h \leq 6$, $0 \leq i \leq 1$ and R is a linear, branched or cyclic alkyl group having 1-8 C 25 atoms or an aryl group, or an alkylamine of formula III:



in which R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl

group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

Particularly preferred examples of the organosilane include aminopropyltriethoxysilane, aminopropyltrimethoxysilane, aminopropylmethyldiethoxysilane and aminopropylmethyldimethoxysilane. Of the trialkylamines, preferred are mono-, di- and trialkylamines, preferably C_{1-3} alkyl groups.

Weak acids such as formic acid, acetic acid, propionic acid or citric acid, can also be employed in the process of the invention. If weak acids and weak bases are employed simultaneously, it is not necessary to maintain an equimolar ratio between acid and base.

In the process of the invention, acid salts which can be employed include, for example, alkali metal hydrogen sulfate or dihydrogen phosphate or aluminum acetate, and basic salts which can be employed include, for example, magnesium hydroxide or alkali metal acetate or alkali metal bicarbonate or alkali metal carbonate.

The reaction medium of the present process is preferably the alcohol corresponding to the alkoxy group of the organosilane employed; methanol, n-propanol, i-propanol, n-butanol, i-butanol, t-butanol and/or 2-methoxyethanol. Ethanol is particularly employed.

Linear perfluoroalkylalkoxysilanes are preferred in the present process such as, for example, perfluorododecyl/perfluorodecyl/perfluorooctyltri-alkoxysilanes, in particular perfluorooctyltriethoxysilane,

and those fluoroalkyl-functional organosilanes which, according to formula I, contain $\text{CF}_3(\text{CF}_2)_7-$, $\text{CF}_3(\text{C}_6\text{H}_4)_6-$, C_6F_5- or $\text{R}^f\text{CH}_2\text{CH}_2(\text{C}=\text{O})-$, where $\text{R}^f = \text{C}_n\text{F}_{2n+1}$ and $n = 2-18$, as the group R^i , for example $\text{F}_{13}\text{C}_6(\text{CH}_2)_2\text{Si}(\text{OC}_2\text{H}_5)_3$. Particularly preferred are

5 tridecafluoro-1,1,2,2-tetrahydrooctyltrimethoxysilane and tridecafluoro-1,1,2,2-tetrahydrooctyltriethoxysilane and 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptafluoro-1,1,2,2-tetrahydrodecyltriethoxysilane and mixtures thereof.

Products prepared by the present process are stable and

10 as a rule exist as clear solutions. The compositions of the invention can be diluted with alcohols in any ratio as desired, and can comprise water in amounts of up to 5% by weight.

The present fluoroalkyl-functional organosiloxanes

15 preferably have a high concentration of silanol functional groups, on the basis of their oligomeric structure, which render them capable, in an outstanding manner, of reaction with substrate surfaces containing hydroxyl groups. Coatings and impregnations with various substrates resulted in

20 excellent oil- and simultaneously water-repellent properties, even after treatment with heat, surfactants and UV, the examples. In appropriate studies on various materials, the present compositions have shown no reduction in efficacy or destabilization even after >6 months.

25 The compositions of the invention or prepared by the present process are used with significant advantages over the products of the prior art. Using the composition of the invention, at the same time a hydrophobizing, oleophobizing

and dirt- and color-repellent action can be achieved in a simple and outstanding manner on the most diverse substrate substances, in particular on glass, for example, plate glass, glass fibers, and glass beads, fillers and pigments, metals, plastics, coatings and paints, textile fibers, including cotton, wood, paper, mineral fibers and mineral building materials such as lime, sandstone, concrete, brick and ceramic. The present compositions can furthermore also be used as release agents, as crosslinking agents, as adhesion promoters, in particular for fluorine polymers, such as, for example, Teflon or coatings based on fluorine polymers, and as additives for paints and coatings. Corresponding processes for surface treatment using compositions of the invention can in general be carried out with little industrial expenditure or expenditure on time.

The present invention, therefore, also relates to the use of the alcohol solvent based fluoroalkyl functional group containing organosiloxane-containing compositions based on alcohol of the invention for hydrophobizing and oleophobizing and simultaneously providing a dirt- and color-repellent treatment of surfaces, of plastics, of metals, of textiles, leather, cellulose and starch products and of mineral building materials, for protecting buildings and facades, for coating glass fibers, for silanizing fillers and pigments, for improving the rheological properties of polymer dispersions and emulsions, and for use as release agents, as crosslinking agents, as adhesion promoters and as additives for paints and coatings.

Having generally described this invention, a further understanding can be obtained by reference to certain specific examples which are provided herein for purposes of illustration only and are not intended to be limiting unless otherwise specified.

Example 1:

A 10 g amount of H₂O (0.56 mol) is mixed with 5 g of HCOOH (pure: 0.11 mol) in a 250 ml glass beaker at room temperature, while stirring, and 160 g of ethanol (3.5 mol) are then added, and a pH of about 3.5-4 being established. Thereafter, 10 g of VPS 8261 (3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluoro-1,1,2,2-tetrahydrooctyltriethoxysilane) are added dropwise (0.02 mol) and the mixture is subsequently stirred for 5 hours with no further change in pH. Thereafter, the material balance is established at a total weight of 1000 g with 915 g of ethanol (19.9 mol).

A clear, colorless solution which has a content of 1.0% by weight of VPS 8261 and a storage stability of at least 6 months is obtained.

Comparison Example:

The procedure is as described in Example 1, but HCOOH is replaced by 1 molar HCl (pH \leq 1). A milky cloud thus formed after 48-72 hours, and thereafter a significant phase separation takes place within 2 weeks. After this period, the composition appears only slightly cloudy, but with a translucent, waxy layer which settles on the base and walls of

containers of both glass and plastic.

Example 2:

5 A 10 g amount of H₂O (0.56 mol) is mixed with 4.0 g of
CH₃COOH (pure: 0.09 mol) in a 250 ml glass beaker at room
temperature, while stirring, and 2.0 g (pure: 0.03 mol) of
isobutylamine is added, a pH of 4.5-5 being established.
Thereafter, the material balance is established with 50 g of
ethanol (1.1 mol). A 5 g amount of
10 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10-heptadecafluoro-1,1,2,2-
tetrahydrodecyltriethoxysilane) is added dropwise (0.008 mol)
and the mixture is subsequently stirred for 5 hours with no
further change in pH. Thereafter, the material balance is
established at a total weight of 500 g with 429 g of ethanol
15 (94.4 mol).

A clear, colorless solution which has a content of 1.0%
by weight of 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-
heptadecafluoro-1,1,2,2-tetrahydrodecyltriethoxysilane and a
storage stability of at least 6 months is obtained.

Example 3:

20 A 25 g amount of H₂O (1.4 mol) is mixed with 5 g of HCOOH
(pure: 0.11 mol) in a 250 ml glass beaker at room temperature,
while stirring, 10 g of DYNASYLAN® 1203 (3-
25 aminopropyltriethoxysilane) are added dropwise (0.07 mol) and
50 g of ethanol (1.1 mol) are then added. A pH of about 8.0
is established. Thereafter, 5 g of VPS 8261
(3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluoro-1,1,2,2-

1
tetrahydrooctyltriethoxysilane) (0.01 mol) are added dropwise
and the mixture is subsequently stirred for 5 hours (no
further change in pH). Thereafter, the material balance is
established at a total weight of 750 g with 655 g of ethanol
5 (8.8 mol).

A clear, colorless solution which has a content of 0.67%
by weight of VPS 8261 and a storage stability of at least 4
months is obtained.

10 Example 4:

COPIED FROM THE PATENT
A 15 g amount of DYNASYLAN® 1203 (3-
aminopropyltriethoxysilane) (0.045 mol) is mixed with 5 g of
VPS 8261 (3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluoro-1,1,2,2-
tetrahydrooctyltriethoxysilane) (0.07 mol) and 50 g of ethanol
15 (1.1 mol) in a 250 ml glass beaker at room temperature.

Thereafter, 25 g of H₂O are added with continued stirring. A
pH of about 9-10 is established. The mixture is subsequently
stirred for a further 5 hours (no further change in pH) and
the material balance is established at a total weight of 500 g
20 with 405 g of ethanol (8.8 mol).

A clear, colorless solution which has a content of 1.0%
by weight of VPS 8261 and a storage stability of at least 4
months is obtained.

25 Example 5:

Using the solutions of Examples 1-4, coatings were
produced on glass sheets, steel sheets, aluminum foils, copper
sheets, cardboard, paper and polyester and impregnations were

produced on cellulose, wood. leather and cotton.

In order to treat these materials, the substrate specimens were immersed in the ready-to-use solutions of Examples 1-4 for about 5 minutes and, after removal, the solvents were removed at 120°C in a drying cabinet over a period of about 30 minutes.

Thereafter, the substrate specimens showed significant hydro- and oleophobic properties. These characteristics are demonstrated, inter alia, by the following properties:

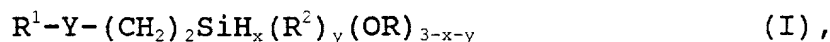
Beading effect of heating oil, silicone oil, water, contact angle > 100°, "anti-graffiti", "anti-soiling", "anti-fouling". Even after storage of the formulations for several months at room temperature, with varying moisture contents and under the influence of daylight, substrates treated with the formulations exhibit these favorable characteristics.

The disclosure of German priority Application No. 196 49 954.2 filed December 3, 1996 is hereby incorporated by reference into the application.

Obviously numerous modifications and variations of the present invention are possible in light of the above teachings. It is, therefore, to be understood that within the scope of the appended claims, the invention may be practiced otherwise than as specifically described herein.

WHAT IS CLAIMED AS NEW AND IS DESIRED TO BE SECURED BY LETTERS
PATENT OF THE UNITED STATES IS:

1. An alcoholic fluoroalkyl-functional group containing
5 organosiloxane based composition prepared by the controlled
hydrolysis of at least one fluoroalkyl-functional group
containing organosilane of formula I:



10 in which R^1 is a mono, oligo- or perfluorinated alkyl
group having 1-9 C atoms or a mono-, oligo- or perfluorinated
aryl group, Y is a CH_2 , O or S group, R^2 and R are each
independently a linear, branched or cyclic alkyl group having
1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or
2, where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ C$
15 over a period of 0.5-24 hours and with thorough mixing in an
alcoholic medium which contains water and a weak mono- or
polybasic acid or a weak base or a weak mono- or polybasic
acid and a weak base or an acid or basic salt, the water
employed and the alkoxysilane employed being in a molar ratio
20 of 2-500:1.

2. The composition as claimed in Claim 1, which has a
pH of 2-12.

3. The composition as claimed in Claim 1, wherein the
alcohol content in the composition is 40-99.999% by weight.

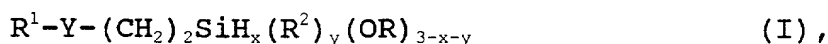
25 4. The composition as claimed of Claim 1, which is free
of chlorine.

5. The composition as claimed in Claim 1, which
contains fluoroalkyl-functional group containing
organosiloxanes in amounts of 0.001-30% by weight, based on

the composition.

6. The composition as claimed in Claim 1, which has a viscosity of less than 10,000 mPa.s.

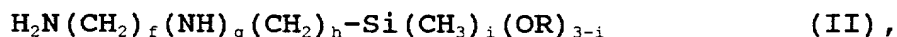
7. A process for the preparation of a fluoroalkyl-functional group containing organosiloxane composition as claimed in Claim 1, which comprises subjecting at least one fluoroalkyl-functional group containing organosilane of formula I:



in wherein R^1 is a mono, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, to controlled hydrolysis with thorough mixing, in an aqueous alcoholic medium which contains a weak mono- or polybasic acid or a weak base or a weak mono- or polybasic acid and a weak base or an acid or basic salt, at a temperature in the range of $0-120^\circ C$ and over a period of 0.5-24 hours, the water and the alkoxysilane being employed in a molar ratio of 2-500:1.

8. The process as claimed in Claim 7, wherein the pH in the reaction medium is adjusted to a value of 2-12.

9. The process as claimed in Claim 7, wherein said weak base is an organosilane of formula II:



in which $0 \leq f \leq 6$, $g=0$ if $f=0$ and $g=1$ if $f>1$, $0 \leq h \leq 6$, $0 \leq i \leq 1$ and R is a linear, branched or cyclic alkyl group having 1-8 C

atoms or an aryl group, or an alkylamine of formula III:



wherein R^3 is a linear, branched or cyclic alkyl group having 1-8 C atoms or a linear, branched or cyclic aminoalkyl group having 1-8 C atoms or an aryl group, $z = 1, 2$ or 3 and groups R^3 are identical or different.

10. The process as claimed in Claim 7, wherein said weak acid is formic acid, acetic acid, propionic acid or citric acid.

11. The process as claimed in Claim 7, wherein said acid salt alkali metal hydrogen sulfate, dihydrogen phosphate or aluminum acetate.

12. The process as claimed in Claim 7, wherein said basic salt is magnesium hydroxide, alkali metal acetate, alkali metal bicarbonate or alkali metal carbonate.

13. The process as claimed in Claim 7, wherein said alcohol is the alcohol corresponding to the alkoxy group of the organosiloxane employed.

14. The process as claimed in Claim 7, wherein said alcohol is methanol, ethanol, n-propanol, i-propanol, n-butanol, i-butanol, t-butanol and/or 2-methoxyethanol.

15. A method of hydrophobizing and oleophobizing and for simultaneously providing a dirt- and color-repellent treatment of surfaces, of plastics, of metals, of textiles, leather, cellulose and starch products, and of mineral building materials, by applying to such the alcoholic fluoroalkyl group containing organosiloxane composition of Claim 1.

16. A method of protecting building and facades,
comprising:

applying the alcoholic fluoroalkyl functional group
containing organosiloxane of Claim 1 to buildings and facades.

5 17. A method for coating glass fibers, comprising:
coating said glass fibers with the alcoholic fluoroalkyl
functional group containing organosiloxane of Claim 1.

18. A method of silanizing fillers and pigments,
comprising:

10 applying the alcoholic fluoroalkyl functional group
containing organosiloxane of Claim 1 to said fillers and
pigments.

19. A method of improving the rheological properties of
polymer dispersions and emulsions, comprising:

15 preparing said dispersions and emulsions with the
alcoholic fluoroalkyl functional group containing
organosiloxane of Claim 1.

20. A method of providing a release layer with release
properties, comprising:

20 incorporating the alcoholic fluoroalkyl functional group
containing organosiloxane of Claim 1 in the release layer.

21. A method of formulating paints and coatings,
comprising:

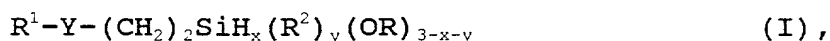
25 incorporating the alcoholic fluoroalkyl functional group
containing organosiloxane of Claim 1 in said paint or coating
material.

22. A method for promoting adhesion of a formulation,
comprising:

incorporating the alcoholic fluoroalkyl functional group
containing organosiloxane of Claim 1 into said formulation.

ABSTRACT OF THE DISCLOSURE

An alcoholic fluoroalkyl-functional group containing organosiloxane based composition prepared is prepared by the controlled hydrolysis of at least one fluoroalkyl-functional group containing organosilane of formula I:



in which R^1 is a mono, oligo- or perfluorinated alkyl group having 1-9 C atoms or a mono-, oligo- or perfluorinated aryl group, Y is a CH_2 , O or S group, R^2 and R are each independently a linear, branched or cyclic alkyl group having 1-8 C atoms or an aryl group and $x = 0, 1$ or 2 and $y = 0, 1$ or 2 , where $(x+y) \leq 2$, at a temperature in the range of $0-120^\circ C$ over a period of 0.5-24 hours and with thorough mixing in an alcoholic medium which contains water and a weak mono- or polybasic acid or a weak base or a weak mono- or polybasic acid and a weak base or an acid or basic salt, the water employed and the alkoxysilane employed being in a molar ratio of 2-500:1.

Declaration, Power Of Attorney and Petition

Page 1 of 3

WE (I) the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FLUOROALKYL-FUNCTIONAL ORGANOSILOXANE-CONTAINING COMPOSITIONS

BASED ON ALCOHOL, A PROCESS FOR THEIR PREPARATION AND THEIR USE

the specification of which

~~XXX~~ is attached hereto.

☐ was filed on _____ as

Application Serial No. _____

and amended on _____.

☐ was filed as PCT international application

Number _____

on _____,

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed	
196 49 954.2	GERMANY	3/December/1996	XXX <input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes	<input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

_____	_____
(Application Number)	(Filing Date)
_____	_____
(Application Number)	(Filing Date)

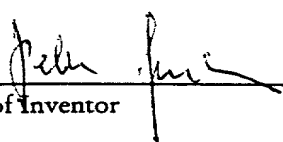
We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.	Filing Date	Status (pending, patented, abandoned)
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint: Norman F. Oblon, Registration Number 24,618; Marvin J. Spivak, Registration Number 24,913; C. Irvin McClelland, Registration Number 21,124; Gregory J. Maier, Registration Number 25,599; Arthur I. Neustadt, Registration Number 24,854; Richard D. Kelly, Registration Number 27,757; James D. Hamilton, Registration Number 28,421; Eckhard H. Kuesters, Registration Number 28,870; Robert T. Pous, Registration Number 29,099; Charles L. Gholz, Registration Number 26,395; Vincent J. Sunderdick, Registration Number 29,004; William E. Beaumont, Registration Number 30,996; Steven B. Kelber, Registration Number 30,073; Robert F. Gnuse, Registration Number 27,295; Jean-Paul Lavalleye, Registration Number 31,451; Timothy R. Schwartz, Registration Number 32,171; Stephen G. Baxter, Registration Number 32,884; Martin M. Zoltick, Registration Number 35,745; Robert W. Hahl, Registration Number 33,893; Richard L. Treanor, Registration Number 36,379; Steven P. Weihrouch, Registration Number 32,829; John T. Goolkasian, Registration Number 26,142; Marc R. Labgold, Registration Number 34,651; William J. Healey, Registration Number 36,160; Richard L. Chinn, Registration Number 34,305; Steven E. Lipman, Registration Number 30,011; Carl E. Schlier, Registration Number 34,426; James J. Kulbaski, Registration Number 34,648; Catherine B. Richardson, Registration Number 39,007; Richard A. Neifeld, Registration Number 35,299; J. Derek Mason, Registration Number 35,270; and Jacques M. Dulin, Registration Number 24,067; our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C., whose Post Office Address is: Fourth Floor, 1755 Jefferson Davis Highway, Arlington, Virginia 22202.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Peter JENKNER
NAME OF FIRST ~~SOLE~~ INVENTOR


Signature of Inventor

19. Aug. 1997
Date

Residence: Friedrich-Ebert-Strasse
8/22, 79618 Rheinfelden,
GERMANY

Citizen of: GERMANY
Post Office Address: (same as above)

Albert-Johannes FRINGS
NAME OF SECOND JOINT INVENTOR

Albert Frings
Signature of Inventor

19. Aug. 1997
Date

Michael HORN
NAME OF THIRD JOINT INVENTOR

Michael Horn
Signature of Inventor

19. Aug. 1997
Date

Eckhard JUST
NAME OF FOURTH JOINT INVENTOR

E. Just
Signature of Inventor

19. Aug. 1997
Date

Jaroslav MONKIEWICZ
NAME OF FIFTH JOINT INVENTOR

Jaroslav Monkiewicz
Signature of Inventor

19. Aug. 1997
Date

Residence: Duererstrasse 50, 79618
Rheinfelden, GERMANY

Citizen of: GERMANY

Post Office Address: (same as above)

Residence: Meisenrain 11, 79618
Rheinfelden, GERMANY

Citizen of: GERMANY

Post Office Address: (same as above)

Residence: Duererstrasse 57, 79618
Rheinfelden, GERMANY

Citizen of: GERMANY

Post Office Address: (same as above)

Residence: Burstelstrasse 2 a,
79618 Rheinfelden, GERMANY

Citizen of: GERMANY

Post Office Address: (same as above)

Burkhard STANDKE
NAME OF SIXTH JOINT INVENTOR


Signature of Inventor

19. Aug. 1997

Date

NAME OF SEVENTH JOINT INVENTOR

Signature of Inventor

Date

NAME OF EIGHTH JOINT INVENTOR

Signature of Inventor

Date

NAME OF NINTH JOINT INVENTOR

Signature of Inventor

Date

Residence: Chrischonastrasse 25,
79540 Loerrach, GERMANY

Citizen of: GERMANY

Post Office Address: (same as above)

Residence:

Citizen of:

Post Office Address:

Residence:

Citizen of:

Post Office Address:

Residence:

Citizen of:

Post Office Address: